UCRL-51879 Part 2

ANALYTICAL PROGRAM — 1975 BIKINI RADIOLOGICAL SURVEY

706756

Mark E. Mount
William L. Robison
Stanley E. Thompson
Keith O. Hamby
Austin L. Prindle
Harris B. Levy

REPOSITORY DOE - FORRESTAL
COLLECTION MARKEY FILES
BOX No. 6 OF 6
FOLDER 2.8 MARSHALLESE ISLANDS #2 (REPORTS ETC.)

November 11, 1976

Prepared for U.S. Energy Research & Development Administration under contract No. W-7405-Eng-48

LAWRENCE LIVERMORE LABORATORY
University of California/Livermore

1014377

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research & Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately-owned rights.

NOTICE

Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Energy Research & Development Administration to the exclusion of others that may be suitable.

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161
Price: Printed Copy $; Microfiche $3.00

Page Range	Domestic Price	Page Range	Domestic Price
001–025	$ 3.50	326–350	10.00
026–050	4.00	351–375	10.50
051–075	4.50	376–400	10.75
076–100	5.00	401–425	11.00
101–125	5.50	426–450	11.75
126–150	6.00	451–475	12.00
151–175	6.75	476–500	12.50
176–200	7.50	501–525	12.75
201–225	7.75	526–550	13.00
226–250	8.00	551–575	13.50
251–275	9.00	576–600	13.75
276–300	9.25	601–up	*
301–325	9.75		

*Add $2.50 for each additional 100 page increment from 601 to 1,000 pages; add $4.50 for each additional 100 page increment over 1,000 pages.

1014378

Distribution Category
UC-11

LAWRENCE LIVERMORE LABORATORY
University of California/Livermore, California/94550

UCRL-51879 Part 2

ANALYTICAL PROGRAM — 1975 BIKINI RADIOLOGICAL SURVEY

Mark E. Mount,* William L. Robison, Stanley E. Thompson,
Keith O. Hamby, Austin L. Prindle, and Harris B. Levy

MS. date: November 11, 1976

*McClellan Central Laboratory, McClellan AFB, California.

1014379

BLANK PAGE

1014380

Contents

Abstract 1

Introduction 1

Initial Processing of Field Samples 3

Soil Samples 3

Vegetation and Animal Samples 5

Gamma Spectrometry 5

Wet-Chemistry Analyses 6

General 6

Chemistry Procedures 8

Measurement Techniques 12

Quality-Control Program 13

Sample Aliquot 13

Sample Homogeneity 15

Data Comparison 15

Measurement of ^{241}Am Concentration — Alpha vs Gamma Detection . . . 15

Wet-Chemistry Analyses of ^{241}Am vs $^{239,240}Pu$ 19

Plutonium-Isotope Results 20

Acknowledgments 26

1014381

BLANK PAGE

1014382

ANALYTICAL PROGRAM — 1975 BIKINI RADIOLOGICAL SURVEY

Abstract

The analytical program for samples of soil, vegetation, and animal tissue collected during the June 1975 field survey of Bikini and Eneu islands is described. The phases of this program are discussed in chronological order: initial processing of samples, gamma spectrometry, and wet chemistry. Included are discussions of quality control programs, reproducibility of measurements, and comparisons of gamma spectrometry with wet chemistry determinations of ^{241}Am. Wet chemistry results are used to examine differences in Pu:Am ratios and Pu-isotope ratios as a function of the type of sample and the location where samples were collected.

Introduction

In June 1975 a field survey was conducted on the islands of Bikini and Eneu within the Bikini Atoll. During this survey, several hundred samples were collected to assess the radiological status of the islands and their suitability for reinhabitation by the Bikini people. Instrumental to the radiological assessment was a thorough and comprehensive program for the analysis of collected samples. Since many facets of the Bikini program were similar to those employed for Enewetak, we used the excellent discussion of the Enewetak analytical program by Hoff et al.[1] as a source document in the preparation of this report. A listing of the samples submitted for analysis is presented in Table 1.

More than 950 samples were collected from Bikini and Eneu Islands during field operations. All samples were processed prior to selection for gamma spectrometry and/or wet chemistry. Of the total samples processed, 624 were counted by gamma spectrometry at LLL on the Ge(Li) detector systems of the Biomedical and Radiochemistry Divisions. Wet-chemistry analyses were performed by the McClellan Central Laboratory (MCL) on 588 of

1. R. W. Hoff, J. W. Meadows, H. D. Wilson, A. L. Prindle, R. Gunnink, and K. O. Hamby, "Analytical Program," *Enewetak Radiological Survey*, U.S. Atomic Energy Commission, Nevada Operations Office, NVO-140, Vol. 1, 426-485, October 1973.

1014383

Table 1. Analytical program for Bikini samples.

Sample type	Total collected	Gamma counting	Wet chemistry
Bikini soil	648	369	333
Eneu soil	167	118	118
Bikini vegetation	96	96	96
Eneu vegetation	31	31	31
Bikini animal	10	10	10

the samples analyzed by gamma spectrometry. All radionuclide concentrations, whether determined by gamma spectrometry or wet chemistry, were reported to a reference time of 1 Jan 1975 (001.000 Z, 75).

All initial processing was conducted at LLL and consisted primarily of drying, homogenizing, and packaging the samples. Soil and vegetation samples were dried by heating in ordinary ovens. Ten samples of pig and chicken tissue collected on Bikini were lyophilized.

Wet-chemistry analyses performed by MCL involved the dissolution of a sample aliquot, chemical separation of the desired elements, and radiation measurement of the elemental samples. In no case was an entire sample consumed in a single dissolution. All vegetation and animal tissue samples submitted for wet chemistry had been analyzed previously by gamma spectrometry. Separate aliquots from each large soil sample were submitted for wet chemistry and for gamma spectroscopy. Wet chemistry was required for certain nuclides that could not be measured by gamma counting; the majority of these nuclides were either alpha or beta emitters.

Discussions of the individual quality control programs are included in the sections dealing with gamma spectrometry and wet chemistry. Reproducibility of measurements was examined by statistically comparing ratios of the individual measurements of a given isotope. The mean value and standard deviation of the ratios were then calculated. The significance of a mean value differing from unity, i.e., indication of possible bias, was tested by calculating the standard error, $s_{\bar{\mu}}$ of the mean $\bar{\mu}$ (logarithmic mean) multiplying $s_{\bar{\mu}}$ by a factor t, which is based upon the 95 percent

1014384

confidence level and is obtained from standard tables, and comparing the value of $t \cdot s_{\bar{\mu}}$ with μ. If the logarithmic mean exceeded $t \cdot s_{\bar{\mu}}$, the observed bias was said to be significant with a 95 percent level of confidence.

Initial Processing of Field Samples

SOIL SAMPLES

Soil samples, by far the largest category, were treated similarly to those samples obtained during the 1972 Enewetak Survey.[1] The treatment consisted of drying, pulverizing, blending, screening, packaging, and preliminary gamma assay. Three separate aliquots were produced from each soil sample: an aluminum "tuna can" containing 300 to 350 g and two vials containing 50 g each.

The soil-processing facility was carefully surveyed for possible radioactive contamination. Air-filter samples and swipe samples taken from the floors were analyzed for ^{60}Co, ^{137}Cs, and $^{239+240}Pu$ content. There was no detectable contamination. The area was considered suitable for initial processing of soils. This monitoring program continued throughout operation of the facility.

Drying ovens designed and built for the Enewetak Survey were used for initial drying of samples at approximately 70°C. Two ovens were constructed of asbestos board with steel shelves inside; two 300-W air heaters blew warm air into each unit, which was equipped with a fan in the vent pipe. Final drying was accomplished in a large commercial drying oven at 150°C.

Samples were ground in a 1-gal paint can using eight 1-in. steel balls. The cover of each can was taped securely; then the entire can was covered with a galvanized-steel jacket that was held in place by two large rubber "O" rings to prevent the lids from falling off during ball milling. A maximum of 48 samples could be milled overnight to provide 15 to 24 h of grinding.

Packaging, weighing, and labeling of samples were performed by hand. All work with finely divided soil was performed in fume hoods. Before each sample was packaged, clean paper was laid out on the hood bench. Care was taken to prevent cross-contamination of samples.

The following is a detailed chronological description of the operations:

- The samples were first unpackaged from the shipping container and logged. The appearance of each sample was noted (e.g., amount of

1014385

organic matter, color, presence of large chunks, etc.).

- The samples were transferred to a disposable aluminum cake pan and covered with aluminum foil. Holes were punched in the top of the foil to permit evaporation.
- The samples were transferred to preliminary drying ovens that were designed to handle about 200 samples. These ovens were set at a temperature of approximately 70°C and operated contiunously. The average residence time per sample was 48 h.
- To assure complete dryness, the samples were placed in a second oven at approximately 150°C; the sample residence time averaged about 3 h.
- The samples were transferred to a 1-gal paint can and dry weights were determined. Sample weights varied from 100 g to 2 kg.
- The samples were milled with eight 1-in steel grinding balls. The sample residence time in the ball mill was between 15 to 24 hr.
- The soil was screened through 2-mm grid, stainless-steel screens to produce a uniform, homogeneous sample for analysis.
- The finely ground soil was prepared for gamma spectrometry and wet chemistry analysis by placing it in two different containers. The gamma-spectrometry samples were placed in tightly sealed "tuna cans" made of 0.25-mm-thick aluminum. The large can was 3.9 cm high, 8.3 cm in diameter, with a cross-sectional area of 53.8 cm^2 and a volume of 210 cm^3. The small can was 3.3 cm high, 6.0 cm in diameter, with a cross-sectional area of 28.5 cm^2 and a volume of 95 cm^3. Soil-sample weights in these cans ranged from 100 to 375 g. Two samples for wet-chemistry analysis, each weighing approximately 50 g, were placed in vials. One of the vials was committed to chemical analysis, and the other held as a backup sample.
- The gamma-spectrometry samples were assayed for gross gamma counts with a 3 × 3-in. NaI (Tl) detector; a 512-channel NaI (Tl) gamma spectrum was measured for those samples that exceeded 100 counts/min. These preliminary NaI (Tl) data guided the scheduling of more precise measurements with Ge(Li) detectors and wet-chemistry procedures.

More than 810 samples were processed in the soil-preparation facility between 4 September 1975 and 10 October 1975 by an average working force of 4.5 people.

1014386

VEGETATION AND ANIMAL SAMPLES

Both plant and animal samples were received frozen with dry ice. The plant samples were spread in stainless steel pans and dried at approximately 80°C for at least 24 h in a forced draft oven until they reached constant weight. The dried plant materials were ground in a Wiley mill with a 2-mm screen, pressed into the aluminum "tuna cans" with a Carver press at about 14,000 psi, and sealed. Two sizes of cans were used, one containing 210 cm^3, the other 95 cm^3. Samples insufficient in volume to fill a small can were packaged in plastic vials. Sample weights were logged for calculation of specific activities.

Coconut meat, because of its high oil content, was not ground but was broken into small chips and pressed into the aluminum cans. Coconut milk was mixed with formaldehyde and canned. Litter samples were sifted through a 3 1/2-mesh screen (5.613-mm openings) before being pressed into cans.

The animal tissue samples were sliced thinly and freeze dried. Skin and bone were removed from muscle tissue. Freeze-dried tissues were cut into small pieces and pressed into "tuna cans" as described above for plant materials. Aliquots for wet chemistry were packed into 30 mm snap top plastic vials.

Gamma Spectrometry

All gamma measurements of Bikini soil, animal, and vegetation samples were made by the Radiochemistry and Biomedical Divisions of LLL. A total of 624 samples were analyzed, 282 by Radiochemistry and 342 by the Biomedical facility. Radiochemistry used several Ge(Li)-diode detector systems with diodes that were 50 cm^3 or more in volume. The Biomedical facility used four Ge(Li) diodes ranging from 7 to 19 cm^3 in volume. Most samples were analyzed for approximately 1000 min, although some of the more active samples were analyzed for 300 to 400 min. All gamma spectra were transferred to magnetic tape for analysis on a CDC-7600 computer using the GAMANAL code.[2] A detailed description of measurement

2. R. Gunnink and J. B. Niday, *Computerized Quantitative Analysis by Gamma Ray Spectrometry, Vols. 1-4,* Lawrence Livermore Laboratory, Rept. UCRL-51061 (1971).

1014387

equipment, calibration procedures, and GAMANAL are given in the Enewetak Radiological Survey report.[1]

Most of the Bikini samples were packaged in 3.3- or 3.9-cm-high aluminum cans with nominal volumes of 95 and 210 cm^3, respectively. Isotopic activities are reported as disintegrations per minute per gram (dpm/g). Eleven nuclides have been observed in Bikini samples: ^{60}Co, ^{102m}Rh, ^{106}Ru, ^{125}Sb, ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{155}Eu, ^{207}Bi, ^{235}U, and ^{241}Am. When these radionuclides were not detected, upper limits were calculated by defining the upper limit photopeak area to be twice the square root of the number of counts observed in the continuum normally occupied by the photopeak.

Thirty-one samples were submitted for comparative measurement to both the Biomedical and Radiochemistry facilities. The results are presented in Table 2. Testing the ratio for bias indicated that there was no significant difference in the results from the two facilities and that on the average, for a series of samples, both would obtain the same result. Statistical variation does, of course, exist in the measurement of any individual sample, but for dose assessment the average value of many samples is the important factor.

In addition to the interfacility comparison, a series of samples originally measured in the Biomedical facility was resubmitted to the facility at a later date for comparison of the analytical results. The data are presented in Table 3. Again there is no indication of any statistical bias in the data.

These comparisons reconfirm the reproductibility of results within a facility and between facilities observed during the course of the analytical work for the 1972 Enewetak survey.[1]

Wet-Chemistry Analyses

GENERAL

Wet-chemistry analyses were required to quantify a number of nuclides that could not be determined by gamma spectrometry. Briefly, this procedure involves the dissolution of a sample in the presence of a known amount of elemental carrier or tracer, chemical separation and purification of the desired element, gravimetric or tracer yielding, and quantification by an appropriate technique. Table 4 presents a list of the measured nuclides, their half-lives, principal radiation, and

1014388

Table 2. Concentrations of ^{137}Cs in selected vegetation and soil samples. Biomedical vs Radiochemistry gamma detection.

Master log number	Biomed dpm/g	Radiochem dpm/g	Biomed / Radiochem
01-0065-01	47.1 ±1.3%	37.7 ±0.8%	1.25 ±1.5%
01-0088-91	26.8 ±1.3	22.2 ±3.5	1.21 ±3.7
01-0111-01	266 ±1.1	210 ±0.9	1.27 ±1.4
01-0134-01	108 ±1.1	104 ±1.2	1.04 ±1.6
01-0157-01	156 ±0.8	152 ±1.2	1.03 ±1.4
01-0226-32	84.0 ±1.1	70.2 ±1.8	1.20 ±2.1
01-0318-95	2.20±9.5	1.86±1.6	1.18 ±9.6
01-0456-32	119 ±1.0	128 ±1.0	0.930±1.4
01-0479-32	126 ±0.9	125 ±1.2	1.01 ±1.5
01-0800-10	1500 ±0.8	1970 ±1.2	0.761±1.4
01-0804-10	1930 ±0.7	1910 ±0.8	1.01 ±1.1
01-0808-10	483 ±2.1	500 ±1.0	0.966±2.3
01-0813-10	77.5 ±1.7	70.2 ±1.6	1.10 ±2.3
01-0816-10	541 ±1.0	554 ±0.8	0.976±1.3
01-0817-10	106 ±1.2	94.2 ±1.1	1.13 ±1.6
01-0821-10	169 ±1.1	140 ±1.1	1.21 ±1.6
01-0822-10	660 ±0.8	686 ±0.9	0.962±1.2
01-0831-10	864 ±0.8	840 ±0.9	1.03 ±1.2
01-0841-10	954 ±1.2	882 ±0.9	1.08 ±1.5
01-0846-10	229 ±0.9	264 ±1.3	0.867±1.6
01-0856-10	1150 ±0.8	1110 ±0.9	1.04 ±1.2
01-0860-10	527 ±0.8	536 ±1.0	0.983±1.3
01-0872-10	271 ±1.9	342 ±0.9	0.792±2.1
06-0928-10	3.59±4.4	5.05±2.9	0.711±5.3
01-1001-10	167 ±1.3	154 ±1.0	1.08 ±1.6
01-1019-10	80.6 ±1.1	71.5 ±1.4	1.13 ±1.8
06-0664-01	28.0 ±1.8	27.8 ±1.8	1.01 ±2.5
06-0709-32	25.0 ±2.3	27.2 ±1.2	0.919±2.6
06-0755-32	28.4 ±2.9	29.0 ±2.2	0.979±3.6
06-0893-92	2.73±5.9	2.35±5.1	1.16 ±7.8
06-0928-10	3.59±4.4	5.05±0.9	0.711±4.5
		Average	1.0±15%

1014389

Table 3. Concentrations of ^{137}Cs in selected soil samples. Duplicate counting for gamma detection.

Master log number	Measurement 1 dpm/g	Measurement 2 dpm/g	Meas 1 / Meas 2
01-0525-71	21.7 ±1.3%	21.2 ±2.7%	1.02 ±3.0%
01-0548-72	4.95±4.8	5.39±3.0	0.918±5.7
01-0617-92	34.4 ±1.1	32.3 ±1.5	1.07 ±1.9
01-0191-32	80.6 ±2.7	83.9 ±1.8	0.961±3.2
01-0212-32	86.6 ±1.2	78.4 ±1.0	1.10 ±1.6
01-0269-32	223 ±1.0	201 ±0.9	1.11 ±1.3
01-0353-91	74.2 ±1.5	73.4 ±1.0	1.01 ±1.8
01-0384-92	70.0 ±1.0	72.8 ±1.0	0.962±1.4
01-0422-32	153 ±1.8	160 ±1.1	0.956±2.1
01-0463-32	297 ±0.9	272 ±0.9	1.09 ±1.3
01-0481-31	475 ±1.8	508 ±0.8	0.935±2.0
01-0561-74	202 ±0.8	182 ±1.7	1.11 ±1.9
		Average	1.02±7.2%

method of measurement. Most of the analyses were for ^{90}Sr and $^{239,240,241}Pu$. Approximately 14 percent of the samples scheduled for wet chemistry were analyzed for ^{241}Am. The primary purpose of the ^{241}Am analyses was for comparison with the gamma-spectrometry results. In the case of some vegetation samples, these analyses fulfilled a secondary role of extending the sensitivity for ^{241}Am detection to lower levels. The remaining nuclides in Table 4 were measured in only a small fraction of the samples to provide an indication of their existing levels.

Analyses for stable iron, calcium, and strontium were performed on a limited number of samples. Table 5 summarizes the wet-chemistry analyses performed by MCL. Samples provided for wet chemistry were 50-g aliquots of finely divided coral, 20- to 100 plus-g aliquots of mulched vegetation, and 50 plus-g aliquots of lyophilized animal tissue.

CHEMISTRY PROCEDURES

Determinations of ^{90}Sr, Pu, ^{55}Fe, and ^{63}Ni were made in a single sample aliquot. A separate aliquot

1014390

Table 4. Nuclides measured in wet-chemistry analyses.

Nuclide	Half-life	Principal radiation	Type of detection
^{55}Fe	2.7 y	5.95-keV x ray	Gamma counting: NaI(Ti), Ge(Li) detectors.
^{63}Ni	92 y	β particle (E_{max} = 65.9 keV)	Liquid scintillation counter.
^{90}Sr	28.5 y	β particle of ^{90}Y daughter (E_{max} = 2.27 MeV)	Beta counting: gas-filled proportional counter.
^{151}Sm	87 y	β particle (E_{max} = 76 keV)	Liquid scintillation counter.
^{238}Pu	87.8 y	5.50-MeV α	Alpha-pulse-height analysis (Frisch-grid chamber, solid state).
^{239}Pu	24,400 y	5.16-MeV α	Mass spectrometry, alpha-pulse-height analysis.
^{240}Pu	6,540 y	5.17-MeV α	Mass spectrometry, alpha pulse height analysis.
^{241}Pu	14.0 y	β particle (E_{max} = 21 keV)	Mass spectrometry.
^{241}Am	433 y	5.49-MeV α	Alpha-pulse-height analysis.
Fe, Ca, Sr	Stable	None	(Atomic absorption).

1014391

Table 5. Summary of wet-chemistry analyses.

Sample type	Number of samples	Nuclides analyzed
Bikini soil	346[a]	^{55}Fe (10), ^{90}Sr (all), ^{238}Pu (30), $^{239,240}Pu$ (all), ^{241}Pu (259), ^{241}Am (47).
Eneu soil	122[b]	^{90}Sr (all), $^{239,240}Pu$ (all), ^{241}Pu (56), ^{241}Am (15).
Bikini vegetation	96	^{55}Fe (4), ^{63}Ni (4), ^{90}Sr (all), ^{151}Sm (4), $^{239,240}Pu$ (all), ^{241}Pu (15), ^{241}Am (20).
Eneu vegetation	31	^{90}Sr (all), $^{239,240}Pu$ (all), ^{241}Am (2).
Bikini animal	10	^{55}Fe (2), ^{63}Ni (2), ^{90}Sr (all), ^{151}Sm (2), $^{239,240}Pu$ (all), ^{241}Sm (3).

[a] Total includes 13 samples where duplicate soil samples were analyzed.

[b] Total includes 4 samples where duplicate soil samples were analyzed.

was used to determine the ^{241}Am and ^{151}Sm concentrations. For coralline soil and animal tissue, these aliquots were nominally 5 g. A smaller aliquot of approximately 3 g was taken from vegetation samples. In all cases, samples were ashed at 950°C for 8 h as the initial step in the chemical dissolution. The MCL chemistry scheme for the determination of ^{90}Sr and Pu from coralline soils, vegetation, and animal tissue is outlined in Table 6.

Iron-55 was isolated by passing the working solution, containing iron carrier, through a Dowex 1-X8 anion column (NO_3^- form), precipitation of $Fe(OH)_3$ with NH_4OH, adsorption and elution from a Dowex 1-X8 column (Cl^- form), and final mounting by electrodeposition. A separate aliquot of each sample was ashed and dissolved for elemental analysis of iron by atomic absorption spectroscopy. These analyses were required to provide corrections to

1014392

Table 6. Chemistry scheme for determination of ^{90}Sr and Pu in coralline soils, vegetation, and animal tissue.

Dissolution	Fire coral, vegetation, or animal tissue at 950°C for 8 h. Add ash to Y carrier and ^{242}Pu tracer.[a] Dissolve with 12M HCl + 5.5M HI.[b] Add HNO_3, boil to oxidize Pu, convert to Cl^-.
Separation	Load on Dowex 1-X8 column from 12M HCl (Pu-Y separation). Wash column with 12M HCl. (Load and wash to Y purification.) Elute Pu with 12M HCl + saturated NH_4I (to Pu purification).
Y Purification	Precipitate $Y(OH)_3$ by adding NH_4OH. (Note Sr-Y separation time.) Wash precipitate with H_2O; dissolve with 16M HNO_3; dilute with H_2O. Precipitate $Y(OH)_3$ by adding NH_4OH; wash precipitate with H_2O. Dissolve in 0.1M HCl. Extract twice with 10% HDEHP[c] in toluene. Back-extract with 3M HCl. Precipitate $Y(OH)_3$ by adding NH_4OH. Wash with H_2O, dissolve with 12M HCl + H_2O, filter. Precipitate Y oxalate by adding saturated oxalic acid, digest. Filter precipitate, dry, fire to Y_2O_3 at 900°C, 2 h. Weigh, count ^{90}Y betas.
Pu Purification	To column eluant add 5M $NH_2OH{\cdot}HCl$, $LaCl_3$ carrier, saturated NH_4I, $ZrO(NO_3)_2$ carrier. Precipitate LaF_3 by adding HF. Dissolve with HNO_3 + H_3BO_3. Precipitate $La(OH)_3$ by adding NH_4OH. Dissolve with 16M HNO_3, boil. Precipitate $La(OH)_3$ by adding NH_4OH. Wash with H_2O; dissolve with 12M HCl + few drops HNO_3. Load on Dowex 1-X8 column. Wash with 12M HCl, 12M HCl-dilute HF, more 12M HCl. Elute Pu with 12M HCl-saturated NH_4I. a. Add 12 drops H_2SO_4; fume to SO_3 evolution. Transfer to plating cell; electroplate (for Pu determination by α-pulse-height analysis). OR b. Transfer to mass spectrometry for filament loading (for Pu determination by mass spectrometric analysis).

[a] ^{242}Pu was used as a tracer for pulse-height analysis (α-PHA) and for mass spectrometry. Note that ^{238}Pu could be determined only in those samples that were assayed via α-PHA.

[b] The addition of HI is necessary to insure equilibration of plutonium tracer with the plutonium in the working solution.

[c] Bis(2-ethylhexyl)orthophosphoric acid.

1014393

the chemical yields for iron originally present in the samples.

Standard chemical procedures were used for the isolation and purification of ^{63}Ni, ^{151}Sm, and ^{241}Am. Gravimetric measurement of the recovered nickel and samarium carriers provided yields for the ^{63}Ni and ^{151}Sm samples. The addition of ^{243}Am tracer was required to determine the yield of the ^{241}Am samples. Nickel was purified by numerous precipitations as nickel dimethylglyoxime. The rare earth, samarium, was separated from americium on a Dowex 50 cation-exchange column by gradient elution with α-hydroxyisobutyric acid.

MEASUREMENT TECHNIQUES

Strontium-90 was determined by beta measurement of the chemically separated 64-h ^{90}Y daughter. Interferences from radiochemical contaminants were identified and eliminated through least-squares analysis of the data. These procedures are rather common for determination of the long-lived ^{90}Sr parent.

Plutonium-239, 240, and 241 were separately quantified via mass spectrometric measurement techniques. Observation of the characteristic mass-to-charge ratio for each isotope provided the means of separation and measurement. To determine the atom concentrations of $^{239,240,241}Pu$ in each sample, ^{242}Pu mass tracer was added during the chemical dissolution. Specific activities were calculated from the measured atom concentrations and appropriate decay constants. Since ^{238}Pu could not be determined mass spectrometrically, alpha pulse height analyses of ^{242}Pu-traced plutonium samples was required. Alpha pulse-height analysis was also essential for the quantification of ^{241}Am. Chemical yields for the americium samples were determined from the ^{243}Am tracer. Quantification of ^{238}Pu and ^{241}Am was accomplished by the ratio of the characteristic alpha peak areas to those of the appropriate tracers.

Thin NaI(Tl) and planar Ge(Li) diode pulse-height-analysis detection systems were used to measure the characteristic 6-keV Mn x ray of ^{55}Fe. All samples were measured by NaI(Tl). Ge(Li) detection systems served to confirm results and extend the sensitivity for ^{55}Fe detection to lower levels. Sixteen samples were analyzed for their ^{55}Fe content.

Nickel-63 and samarium-151 were determined by liquid scintillation counting at LLL with a Packard Tri-Carb spectrometer.

Errors reported with each result represent the measurement uncertainty and are based primarily on counting

1014394

statistics. For those nuclides with multiple sample determinations, the reported results and errors are those of the simple average.

Quality-Control Program

SAMPLE ALIQUOT

The usual MCL quality control program was expanded to examine the validity of wet-chemistry analysis of small sample (3 to 5 g) aliquots. Specifically questioned was whether the small aliquots were representative of the larger sample. For well-homogenized samples, the small aliquots were known to be representative.

Twenty-seven samples (5 vegetation and 22 soils) were selected for carrier-free dissolution of 25-g aliquots. The term carrier-free describes working solutions obtained by the dissolution of sample in the absence of appropriate carriers and tracers. The major difference between the carrier-free and standard carrier dissolutions was the absence or presence of the carriers and tracers in the working solution. Processing of ^{90}Sr, Pu and ^{241}Am samples from the carrier-free solution was accomplished by adding Y carrier and ^{242}Pu mass tracer to one aliquot and ^{243}Am to another, achieving isotopic exchange in the solution, and proceeding with the standard methods for separation and purification.

Results from dissolution of the 25-g aliquots are compared in Table 7 with those from the smaller aliquots. The ratio of results from carrier to carrier-free dissolutions is given for each of the atom ratios and isotopic concentrations. Errors reported with each entry result from propagation of uncertainties in the individual measurements. Soil samples from both Bikini and Eneu show excellent reproducibility in all major isotopes. Past experiences in mass spectrometric measurement of minor isotopes such as ^{241}Pu would lead one to expect an even greater spread than that observed. The fact that the results are reproducible to within 10 percent is quite encouraging. For the soils there is no indication of a statistically significant bias resulting from the use of small (5 g) sample aliquots.

Comparison of the measurements of samples of vegetation indicate the possibility of a slight bias in the dterminations of ^{90}Sr and ^{239,240}Pu; however, these biases are not significant at the 95 percent confidence level. There is a definite indication of significant

1014395

Table 7. Comparison of radiochemical results from carrier and carrier-free dissolutions. (Ratios are carrier to carrier-free).[a]

Master log number	$^{240:239}Pu$ atom ratio	$^{241:239}Pu$ atom ratio	^{239}Pu dpm/g	$^{239+240}Pu$ dpm/g	^{241}Pu dpm/g	^{241}Am dpm/g	^{90}Sr dpm/g
			Bikini soil				
01-0003-92	1.02 ±0.8%	1.22 ±2.2%	1.04 ±0.8%	1.05 ±0.8%	1.27 ±2.4%	0.959±2.0%	0.985±0.9%
01-0055-90	0.01 ±1.9	1.07 ±5.7	0.996±1.4	0.998±1.4	1.07 ±5.8	1.01 ±1.9	1.03 ±0.7
01-0056-91	0.999±1.2	1.05 ±2.7	1.02 ±1.0	1.02 ±0.9	1.07 ±2.9	1.03 ±3.1	0.959±0.7
01-0057-92	1.00 ±1.8	1.07 ±5.6	0.986±4.0	0.986±3.0	1.06 ±6.9	1.03 ±3.2	0.934±0.6
01-0076-92	0.993±0.5	0.876±4.3	0.996±0.6	0.992±0.5	0.872±4.3	0.874±7.5	1.03 ±0.6
01-0111-01	0.992±0.4	1.04 ±3.4	1.02 ±0.6	1.02 ±0.5	1.06 ±3.4	0.973±1.5	1.04 ±1.2
01-0118-01	1.01 ±0.2	0.996±0.6	0.972±0.5	0.976±0.4	0.968±0.8	1.01 ±1.9	1.02 ±0.6
01-0119-90	1.00 ±1.1	0.955±3.0	1.02 ±0.9	1.02 ±0.9	0.972±3.1	0.956±6.7	1.03 ±0.8
01-0121-92	1.01 ±0.5	———	1.00 ±0.6	1.01 ±0.5	———	0.966±1.5	1.02 ±0.8
01-0288-92	0.992±0.4	0.813±5.7	0.981±0.6	0.976±0.5	0.797±5.8	1.07 ±3.8	0.994±0.8
01-0331-92	1.00 ±0.6	———	0.973±1.1	0.974±0.9	———	0.969±1.7	1.00 ±0.6
01-0341-90	1.01 ±0.6	0.999±2.5	0.970±0.9	0.975±0.7	0.970±2.6	0.921±3.1	0.989±0.5
01-0352-90	1.00 ±1.8	1.07 ±6.2	1.00 ±1.3	1.01 ±1.3	1.08 ±6.4	0.980±1.9	1.00 ±0.7
01-0354-92	1.02 ±1.6	———	1.05 ±1.3	1.07 ±1.2	———	1.25 ±3.3	1.00 ±1.0
01-0384-92	1.01 ±1.0	0.973±1.5	1.00 ±0.8	1.00 ±0.8	0.972±1.7	0.978±2.8	1.03 ±1.2
Average	1.00±9.9%	1.0±10%	1.00±2.4%	1.01±2.8%	1.0±12%	0.998±8.4%	1.00±2.9%
			Eneu soil				
06-0707-32	0.982±2.6%	———	0.998±1.3%	0.989±1.6%	———	0.994±3.4%	1.04 ±1.5%
06-0708-32	1.01 ±0.8	———	0.975±0.9	0.979±0.8	———	1.02 ±3.1	0.988±1.2
06-0719-32	0.974±2.0	———	1.00 ±1.7	0.988±1.6	———	1.05 ±3.8	1.06 ±1.9
06-0722-32	1.03 ±0.9	———	0.897±0.7	0.910±0.7	———	0.990±5.4	1.01 ±3.7
06-0732-32	0.981±1.2%	———	1.01 ±1.1%	1.01 ±1.0%	———	1.16 ±6.0%	1.04 ±2.3%
06-0936-94	0.994±0.8	1.06 ±4.3%	0.970±1.0	0.967±0.8	1.03 ±4.4%	1.01 ±4.4	0.990±1.1
06-0950-73	0.998±1.3	———	0.972±1.0	0.971±1.0	———	1.06 ±2.8	0.943±1.3
Average	0.996±2.0%	———	0.975±3.9%	0.973±3.2%	———	1.04±5.7%	1.01±4.0%
			Bikini vegetation				
01-0639-10	0.995±2.2%	———	1.07 ±1.3%	1.07 ±1.5%	———	0.866±6.3%	1.09 ±1.8%
01-0641-10	0.973±1.3	———	0.996±0.9	0.982±1.0	———	0.767±5.6	0.984±0.5
01-0803-10	1.01 ±1.0	———	0.766±0.7	0.769±0.7	———	0.785±5.6	0.948±1.0
01-0829-10	0.963±2.9	———	0.934±2.5	0.915±2.3	———	0.859±6.3	1.06 ±1.0
01-0850-10	1.00 ±1.4	0.992±4.7%	0.876±2.8	0.877±2.9	0.894±4.5%	0.929±4.9	1.17 ±1.2
Average	0.988±2.0%	———	0.93±12%	0.92±12%	———	0.841±7.8%	1.05±8.4%

[a]All results are reported to a reference time of 1 January 1975 (001.000 Z, 75).

1014396

bias in the determination of ^{241}Am. Although the number of samples compared is a relatively small fraction of the total analyzed, it appears that the standard vegetation aliquots (3 g) may underestimate the $^{239,240}Pu$ concentrations by 7 percent and the ^{241}Am concentrations by as much as 16 percent. Since the compared samples were either litter or roots, sample inhomogeneity is the likely source of these apparent biases. However, it is also possible that these biases may be indicative of an error resulting from surface contamination of vegetation so that the results may not be a true measure of the isotopic uptake by the plant.

SAMPLE HOMOGENEITY

MCL received duplicate samples from 17 large-volume soil specimens. The question to be answered was whether separate samples from a large specimen of finely divided soil could give reproducible results. The standard carrier dissolution was used to process these samples. Analyses were primarily for ^{90}Sr and Pu. Americium-241 was determined in three of the samples. Measurement results are compared in Table 8. The ratio of A to B samples is presented for each of the measured atom ratios and isotopic concentrations. Quoted statistics are derived from the propagation of errors. Sample homogeneity is evidenced by the excellent agreement in the $^{239,240}Pu$ and ^{90}Sr concentrations from both Bikini and Eneu. Results for ^{241}Pu exhibit some spread among the individual data points but are reproducible to within 11 percent at a mean of unity. This spread is but another example of the inherent difficulty of minor isotopic measurement by mass spectrometry. The ^{241}Am comparison, though limited to three points, also shows no significant bias. Comparison of these samples indicates that separate aliquots of soil can be expected to show reproducible results to within the errors presented in Table 8.

Data Comparison

MEASUREMENT OF ^{241}AM CONCENTRATION — ALPHA VS GAMMA DETECTION

Approximately 13 percent of the 624 samples assayed by gamma spectrometry were selected for ^{241}Am wet-chemistry analysis. In each of the 624 samples of soil, vegetation, and animal tissue, ^{241}Am was quantified through either a positive gamma

1014397

Table 8. Comparison of radiochemical results from separate samples of a large-volume soil specimen. (Ratios are A to B sample aliquot).[a]

Master log number	$^{240:239}Pu$ atom ratio	$^{241:239}Pu$ atom ratio	^{239}Pu dpm/g	$^{239+240}Pu$ dpm/g	^{241}Pu dpm/g	^{241}Am dpm/g	^{90}Sr dpm/g
				Bikini soil			
01-0065-01	1.01 ±0.4%	1.21 ±2.1%	1.02 ±0.8%	1.02 ±0.6%	1.23 ±2.2%	———	0.968±1.1%
01-0088-91	1.01 ±0.9	0.860±9.3	1.03 ±0.9	1.03 ±0.8	0.883±9.4	0.909±8.6	0.990±1.7
01-0111-01	0.998±0.6	1.09 ±2.8	1.00 ±1.6	1.00 ±1.3	1.10 ±4.4	0.987±2.1	1.03 ±4.0
01-0134-32	0.985±2.5	0.90 ±15	0.927±4.3	0.920±3.3	0.845±6.5	———	0.950±4.5
01-0157-32	1.01 ±0.3	1.03 ±0.9	1.00 ±0.5	1.01 ±0.4	1.03 ±1.1	———	1.00 ±0.7
01-0180-32	0.997±1.1	0.930±4.8	1.00 ±0.8	0.998±0.9	0.930±4.9	———	9.998±2.4
01-0226-32	1.01 ±0.4	1.01 ±0.8	0.995±0.5	0.999±0.4	1.01 ±1.0	———	1.03 ±2.4
01-0318-95	0.992±2.2	———	0.970±3.3	0.965±2.7	———	———	0.985±2.0
01-0341-90	0.998±0.8	0.955±1.3	1.04 ±2.2	1.04 ±1.9	0.993±2.6	1.08 ±6.4	1.04 ±0.8
01-0387-95	0.984±2.2	0.99 ±28	1.06 ±3.6	1.05 ±2.4	1.1 ±28	———	0.965±2.6
01-0525-71	1.01 ±0.8	1.16 ±7.4	0.956±1.3	0.959±1.0	1.11 ±7.5		0.951±0.9
01-0548-72	0.984±3.5	———	1.07 ±6.1	1.06 ±4.2	———	———	1.03 ±2.6
01-0617-92	1.01 ±1.2	1.06 ±4.4	1.03 ±0.9	1.04 ±0.9	1.09 ±4.5	———	1.02 ±1.4
Average	1.00±1.1%	1.0±11%	1.01±4.0%	1.01±4.0%	1.0±11%	0.992±8.6%	0.997±3.2%
				Eneu soil			
06-0709-32	0.994±1.1%	0.91 ±11%	1.01 ±0.9%	1.01 ±0.9%	0.92 ±11%	———	1.01 ±1.9%
06-0755-32	0.978±3.8	———	1.05 ±2.4	1.04 ±2.6	———	———	1.08 ±6.5
06-0893-92	1.02 ±1.5	———	0.961±1.4	0.971±1.3	———	———	0.988±6.3
Average	0.997±2.1%	———	1.01±4.4%	1.01±3.4%	———	———	1.03±4.7%

[a] All results are reported to a reference time of 1 January 1975 (001.00 Z, 75).

signal or calculation of a detection limit. Wet-chemistry measurements served two purposes: to permit comparison of two different methods for measuring ^{241}Am, and for other samples, to provide greater sensitivity for detecting ^{241}Am than available from routine gamma measurement.

Comparison data for 52 soil samples are presented in Table 9. The mean value for the MCL:LLL ratio is 1.2 ± 16%, with evidence for significant bias. For soil samples, wet chemistry is expected to assess the ^{241}Am concentration more accurately. Uncertainties in self-absorption corrections because of voids resulting from settling of the soil in the can limit the accuracy of ^{241}Am assessment via gamma spectrometry. Thus, it appears that the ^{241}Am data for soil samples reported via gamma spectrometry may be systematically low by about 20 percent. Even so, the effect of a 20 percent bias will be negligible on the estimated external dose since ^{241}Am contributed a very small fraction to the total.

1014398

Table 9. Comparison of alpha-pulse height analysis (α-PHA) and gamma-spectrometric analyses for ^{241}Am in separate batches of soil (MCL vs LLL).[a]

Master log number	α-PHA (MCL) dpm/g	Gamma spectrometry (LLL)dpm/g	MCL:LLL
01-0001-90	9.26 ±0.7%	6.76± 8.9%	1.37± 8.9%
01-0002-91	7.88 ± 1.1	6.6 ±11	1.2 ±11
01-0003-92	11.4 ± 2.9	10.1 ± 8.7	1.13± 9.2
01-0011-90	6.98 ± 4.8	4.9 ± 1.8	1.4 ±19
01-0012-91	4.39 ± 1.7	3.9 ±16	1.1 ±17
01-0045-90	1.56 ± 2.4	1.2 ±31	1.3 ±31
01-0046-91	4.32 ± 3.1	3.29± 4.9	1.31± 5.8
01-0047-92	1.2 ±13	0.98±15	1.2 ±20
01-0055-90	8.04 ± 0.9	6.85± 6.6	1.17± 6.7
01-0056-91	10.6 ± 1.7	8.36± 9.9	1.3 ±10
01-0074-90	5.23 ± 3.0	5.0 ±14	1.0 ±14
01-0075-91	19.5 ± 1.6	15.0 ± 4.2	1.30± 4.5
01-0076-92	27.7 ± 9.5	17.5 ± 9.1	1.6 ±13
01-0086-01	9.67 ± 8.7	10.5 ± 7.8	0.92±12
01-0087-90	2.06 ± 9.2	1.98± 5.6	1.0 ±11
01-0088-91	1.46 ± 8.4	1.3 ±12	1.1 ±15
01-0110-01	11.3 ± 5.8	11.4 ± 7.3	0.991±9.3
01-0111-01	19.3 ± 1.9	17 ±20	1.1 ±20
01-0112-01	23.5 ± 0.8	16.2± 6.8	1.45± 6.8
01-0116-01	18.0 ± 3.4	12.9± 8.5	1.40± 9.2
01-0118-01	26.4 ± 1.0	19.0 ± 5.8	1.39± 5.9
01-0119-90	5.81 ± 3.2	3.9 ±12	1.5 ±12
01-0120-91	8.97 ± 3.4	8.14± 7.6	1.10± 8.3
01-0121-92	14.5 ± 2.5	10.4 ± 5.1	1.39± 5.7
01-0273-90	14.8 ± 3.9	11.4 ± 8.8	1.30± 9.6
01-0274-91	1.20 ± 3.4	1.2 ±25	1.0 ±25
01-0275-92	0.884± 3.2	0.95±30	0.93±30
01-0286-90	7.12 ± 7.0	5.6 ±16	1.3 ±17
01-0287-91	13.4 ± 6.8	11 ±12	1.2 ±14
01-0288-92	21.7 ± 4.5	16.2 ± 7.4	1.34± 8.7

1014399

Table 9. (Continued).

Master log number	α-PHA (MCL) dpm/g	Gamma spectrometry (LLL) dpm/g	MCL:LLL
01-0329-90	13.6 ± 2.5%	10.1 ± 9.1%	1.35± 9.4%
01-0330-91	21.5 ± 7.6	14.0 ± 7.9	1.5 ±11
01-0331-92	41.0 ± 2.2	27.9 ± 6.1	1.47± 6.5
01-0341-90	50.8 ± 5.6	37.9 ± 8.7	1.3 ±10
01-0342-91	7.62 ± 1.8	6.0 ±10	1.3 ±11
01-0343-92	1.02 ± 7.3	1.0 ±50	1.0 ±51
01-0352-90	3.02 ± 1.4	3.0 ±29	1.0 ±29
01-0353-91	1.77 ± 1.8	1.5 ±19	1.2 ±19
01-0382-90	2.74 ± 2.0	2.4 ±32	1.1 ±32
01-0383-91	3.67 ± 1.7	3.6 ±11	1.0 ±11
01-0384-91	4.85 ± 1.6	4.6 ±14	1.1 ±14
06-0707-32	4.07 ± 1.3	4.06± 9.8	1.00± 9.9
06-0718-32	3.30 ± 8.1	2.86± 8.7	1.2 ±12
06-0719-32	2.47 ± 3.7	2.0 ±11	1.2 ±12
06-0722-32	0.811± 2.4	0.74±13	1.1 ±13
06-0752-32	3.2 ±10	2.4 ±13	1.3 ±16
06-0740-32	0.764± 5.1	0.42±43	1.8 ±43
06-0752-32	2.42 ± 4.7	2.90± 6.2	0.834±7.8
06-0758-32	3.78 ± 5.1	2.98± 6.4	1.27± 8.2
06-0765-32	0.72 ±10	0.62±13	1.2 ±16
06-0936-94	6.69 ± 1.2	5.35± 5.4	1.25± 5.5
06-0950-73	4.78 ± 4.1	3.37± 9.5	1.4 ±10
		Average	1.2±16%

[a] All results are reported to a reference time of 1 January 1975 (001.000Z, 75).

1014400

Table 10 presents a comparison of all vegetation samples that showed positive ^{241}Am via gamma spectrometry. This limited data set of five samples exhibits a mean MCL:LLL ratio of 0.95 ± 22%, with no evidence for significant bias. Wet-chemistry results are the simple averages of the individual determinations by carrier and carrier-free dissolution procedures. As indicated earlier in the section on quality control, wet-chemistry determinations of ^{241}Am in vegetation may be systematically low. In addition, the large uncertainties in the individual gamma measurements provide for a very broad range of possible ratios. Thus, there is no reason to conclude that there is any significant difference between wet chemistry and gamma spectrometry of ^{241}Am in vegetation.

In all other cases wet chemistry provided a more sensitive measure of ^{241}Am concentration than did gamma spectrometry. For vegetation samples, increases in sensitivity were in the range of 2.1- to 637-fold. Animal samples exhibited increases in the range of 1.2- to 7.4-fold.

WET-CHEMISTRY ANALYSES OF ^{241}Am VS $^{239,240}Pu$

Results for ^{241}Am and $^{239,240}Pu$ were compared in those samples selected for wet chemistry. Concentration ratios of ^{241}Am to ^{239}Pu and to $^{239+240}Pu$ were calculated. The purposes of these computations were to examine any differences between sample types (soil vs vegetation) and sampling location (Bikini vs Eneu), and to determine mean ratios

Table 10. Comparison of α-PHA and gamma-spectrometric analyses for ^{241}Am in vegetation (MCL vs LLL).[a]

Master log number	α-PHA (MCL) dpm/g	Gamma spectrometry (LLL), dpm/g	MCL:LLL
01-0639-10	0.44±10%	0.34±30%	1.3 ±32%
01-0641-10	0.75±19	0.91±55	0.82±58
01-0803-10	4.9 ±17	6.1 ±20	0.80±26
01-0829-10	0.43±11	0.51±56	0.84±57
01-0850-10	1.67± 5.2	1.6 ±30	1.0 ±30
		Average	0.95±22%

[a] All results are reported to a reference time of 1 January 1975 (001.000Z, 75).

1014401

for the calculation of wet-chemistry-equivalent ^{241}Am concentrations. Comparisons of the Bikini soil, Eneu soil, and Bikini vegetation are presented in Tables 11, 12, and 13, respectively. Only those samples giving positive ^{241}Am and $^{239,240}Pu$ signals have been included.

From Table 11 it is apparent that the Bikini soils exhibit quite consistent ^{241}Am:Pu ratios regardless of the profile depth. In fact, the agreement among the mean ratios for the various profiles is rather remarkable. Results indicate that the average dpm ratios of ^{241}Am:^{239}Pu and ^{241}Am:$^{239+240}Pu$ on the island are quite specific and are 1.17 ± 8.1% and 0.550 ± 8.1%, respectively. As evidenced in Table 12, there is also excellent agreement among the Eneu soil samples. Although the total number of samples is considerably less, there is no appreciable variation with profile depth. For Eneu Island soil samples, the ^{241}Am:^{239}Pu and ^{241}Am:$^{239+240}Pu$ dpm ratios are 1.08 ± 3.0% and 0.512 ± 3.6%, respectively. Statistically, there is no difference between the ^{241}Am to Pu concentration ratios of these two islands. Bikini vegetation ratios in Table 13 are in reasonable statistical agreement as indicated by the fact that the average dpm ratios exhibit lower deviations than any of the individual determinations. Although the average ^{241}Am:^{239}Pu and ^{241}Am:$^{239+240}Pu$ dpm ratios of 1.11 ± 8.1% and 0.512 ± 7.9% are different than those in the Bikini soils when compared on an absolute basis, the deviations associated with the individual determinations indicate that the soil and vegetation ratios are the same.

PLUTONIUM-ISOTOPE RESULTS

Mean isotopic atom ratios of ^{240}Pu and ^{241}Pu to ^{239}Pu have been calculated from the individual mass spectrometric results. These ratios from the various sample types and sampling locations are presented in Table 14. Also included are the activity ratios as determined from the mean atom ratios and half-lives listed in Table 4. Soil results from the two islands are indistinguishable. Statistically, the soil and vegetation results are in agreement. The absolute differences between the ^{240}Pu:^{239}Pu soil and vegetation results show the difficulties associated with the measurement of minor isotope in samples with low concentrations. Extensive experience in mass spectrometry has shown that for low level samples such as vegetation, measurement of minor isotopes will be biased higher than the actual concentrations. To construe that the differences in soil and vegetation are an indication of fractionation would be erroneous.

1014402

Table 11. Wet-chemistry results of ^{241}Am vs Pu in Bikini soil.[a]

Master log number	^{239}Pu dpm/g	$^{239+240}Pu$ dpm/g	^{241}Am dpm/g	^{241}Am:^{239}Pu dpm ratio	^{239}Am:$^{239+240}Pu$ dpm ratio
			Profile 000-005		
01-0001-90	7.53±0.8%	15.7 ±0.7%	9.26±0.7%	1.23 ±1.0%	0.590±1.0%
01-0011-90	5.64±0.9	11.9 ±0.6	6.98±4.8	1.24 ±4.9	0.588±4.9
01-0045-90	1.30±1.8	2.82±2.0	1.56±2.4	1.21 ±3.0	0.554±3.1
01-0055-90	6.98±0.7	15.0 ±0.7	8.04±0.9	1.15 ±1.1	0.536±1.1
01-0074-90	4.33±0.5	9.23±0.4	5.23±3.0	1.21 ±3.0	0.566±3.0
01-0087-90	2.29±0.8	4.82±0.7	2.06±9.2	0.900±9.2	0.427±9.2
01-0119-90	4.81±1.3	10.2 ±1.4	5.81±3.2	1.21 ±3.5	0.570±3.5
01-0273-90	12.2 ±1.0	26.0 ±1.1	14.8 ±3.9	1.21 ±4.0	0.570±4.0
01-0286-90	5.75±0.9	12.3 ±0.9	7.12±7.0	1.24 ±7.0	0.580±7.0
01-0329-90	12.4 ±1.1	26.1 ±1.0	13.6 ±2.5	1.10 ±2.7	0.520±2.6
01-0341-90	39.5 ±2.1	84.1 ±1.8	50.8 ±5.6	1.29 ±6.0	0.604±5.9
01-0352-90	2.64±0.6	5.58±0.6	3.02±1.4	1.14 ±1.5	0.541±1.5
01-0382-90	2.47±0.5	5.25±0.5	2.74±2.0	1.11 ±2.1	0.523±2.1
01-9341-90	38.0 ±0.7	81.0 ±0.5	47.1 ±2.8	1.24 ±2.8	0.582±2.8
			Average	1.18 8.2%	0.554±8.0%
			Profile 005-010		
01-0002-91	6.91±0.5%	14.6 ±0.4%	7.88±1.2%	1.14 ±1.2%	0.538±1.2%
01-0012-91	3.87±0.6	8.33±0.8	4.39±1.7	1.13 ±1.8	0.528±1.9
01-0046-91	3.56±0.5	7.66±0.4	4.32±3.1	1.21 ±3.1	0.563±3.1
01-0056-91	8.89±1.2	18.7 ±1.2	10.6 ±1.7	1.19 ±2.1	0.567±2.1
01-0076-91	15.8 ±0.5	33.6 ±0.5	19.5 ±1.6	1.23 ±1.7	0.579±1.7
01-0088-91	1.46±0.6	3.09±0.6	1.46±8.4	0.998±8.4	0.474±8.4
01-0111-01	16.6 ±1.6	35.0 ±1.3	19.3 ±1.9	1.16 ±2.5	0.551±2.3
01-0120-91	7.71±0.6	16.3 ±0.5	8.97±3.4	1.16 ±3.5	0.550±3.4
01-0274-91	1.05±0.5	2.38±0.6	1.20±3.4	1.14 ±3.5	0.505±3.5
01-0287-91	11.8 ±0.4	25.1 ±0.3	13.4 ±6.8	1.14 ±6.8	0.536±6.8
01-0330-91	15.2 ±0.5	32.1 ±0.4	21.5 ±7.6	1.41 ±7.6	0.669±7.6
01-0342-91	5.52±0.6	11.9 ±0.5	7.62±1.8	1.38 ±1.9	0.639±1.8
01-0353-91	1.62±1.4	3.49±1.3	1.77±1.8	1.10 ±2.2	0.508±2.2

1014403

Table 11. (Continued).

Master log number	^{239}Pu dpm/g	$^{239+240}Pu$ dpm/g	^{241}Am dpm/g	$^{241}Am:^{239}Pu$ dpm ratio	$^{241}Am:^{239+240}Pu$ dpm ratio
01-0383-91	3.20 ±3.2%	7.06±3.8%	3.67 ±1.7%	1.15 ±3.6%	0.520±4.2%
01-9088-91	1.43 ±0.6	2.99±0.5	1.61 ±2.0	1.13 ±2.1	0.538±2.1
01-9111-91	16.6 ±0.4	34.9 ±0.3	19.6 ±0.9	1.18 ±1.0	0.561±1.0
			Average	1.18±8.4%	0.552±8.7%
		Profile 010-015			
01-0003-92	9.53 ±2.9%	10.2 ±3.5%	11.4 ±2.9%	1.20± 4.1%	0.564± 4.5
01-0047-92	1.14 ±1.5	2.41±1.3	1.2 ±13	1.0 ±13	0.49 ±13
01-0076-92	22.9 ±0.3	48.6 ±0.5	27.7 ±9.5	1.21± 9.5	0.570± 9.5
01-0121-92	11.9 ±0.3	25.2 ±0.4	14.5 ±2.5	1.22± 2.5	0.575± 2.5
01-0275-92	0.738±0.9	1.72±1.0	0.884±3.2	1.20± 3.4	0.514± 3.4
01-0288-92	18.0 ±1.4	38.2 ±1.7	21.7 ±4.5	1.21± 4.7	0.568± 4.8
01-0331-92	30.7 ±1.9	63.8 ±1.9	41.0 ±2.2	1.34± 2.9	0.643± 2.9
01-0343-92	0.899±0.8	1.98±1.0	1.02 ±7.3	1.13± 7.3	0.512± 7.4
01-0354-92	0.588±3.8	1.25±4.7	0.69 ±16	1.2 ±16	0.55 ±17
01-0384-92	4.25 ±0.4	8.97±0.4	4.85 ±1.6	1.14± 1.6	0.541± 1.6
			Average	1.18±7.3%	0.553±7.8%
		Surface			
01-0110-01	9.92±0.5	21.0±0.5	11.3±5.8	1.14±5.8	0.539±5.8
		Profile 015-020			
01-0112-01	20.4±0.4	42.5±0.3	23.5±0.8	1.16±0.9	0.554±0.8
		Profile 015-025			
01-0057-92	14.7±1.0	31.1±1.0	17.5±2.1	1.19±2.3	0.563±2.3
		Profile 105-115			
01-0086-01	9.04±0.6	18.8±0.4	9.67±8.7	1.07±8.7	0.514±8.7
		Soil around roots (Papaya No. 1 House 24)			
01-0116-01	16.6±0.5	34.7±0.4	18.0±3.4	1.09±3.4	0.519±3.4

1014404

Table 11. (Continued)

Master log number	^{239}Pu dpm/g	$^{239+240}Pu$ dpm/g	^{241}Am dpm/g	^{241}Am:^{239}Pu dpm ratio	^{241}Am:$^{239+240}Pu$ dpm ratio
	Soil under roots (Papaya No. 1 House 24)				
01-0117-01	0.0734±0.9	0.153±1.5	0.073±15	1.0±15	0.48±15
	Soil under plastic (Papaya No. 1 House 24)				
01-0118-01	20.0±2.0	41.9±1.7	26.4±1.0	1.32±2.2	0.630±2.0
			Overall Soil Average	1.17±8.1%	0.550±8.1%

1014405

Table 12. Wet-chemistry results of ^{241}Am vs Pu in Eneu soil.[a]

Master log number	^{239}Pu dpm/g	$^{239+240}Pu$ dpm/g	^{241}Am dpm/g	$\frac{^{241}Am}{^{239}Pu}$ dpm ratio	$\frac{^{241}Am}{^{239+240}Pu}$ dpm ratio
		0-15 cm soil sample			
06-0707-32	3.62 ±0.6%	7.63 ±0.8%	4.07 ± 1.3%	1.12± 1.4%	0.533± 1.5%
06-0708-32	8.02 ±1.8	17.1 ±1.5	9.02 ± 1.3	1.12± 2.2	0.527± 2.0
06-0718-32	3.05 ±0.4	6.38 ±0.4	3.03 ± 8.1	1.08± 8.1	0.517± 8.1
06-0719-32	2.34 ±0.5	4.84 ±0.9	2.47 ± 3.7	1.05± 3.7	0.510± 3.8
06-0722-32	0.756 ±7.7	1.60 ±6.7	0.811 ± 2.4	1.07± 8.1	0.508± 7.1
06-0727-32	0.0854±2.7	0.190±4.4	0.0895± 9.4	1.05± 9.8	0.47 ±10
06-0732-32	2.89 ±1.1	6.03 ±0.4	3.2 ±10	1.1 ±10	0.53 ±10
06-0740-32	0.725 ±0.5	1.54 ±0.5	0.764 ± 5.1	1.05± 5.2	0.498± 5.2
06-0752-32	2.28 ±0.6	4.72 ±0.6	2.42 ± 4.7	1.06± 4.7	0.512± 4.7
06-0753-32	2.88 ±0.9	5.99 ±0.8	2.94 ± 3.0	1.02± 3.2	0.491± 3.1
06-0758-32	3.58 ±1.1	7.52 ±1.1	3.78 ± 5.1	1.06± 5.2	0.503± 5.2
06-0765-32	0.680 ±1.0	1.40 ±1.0	0.72 ±10	1.1 ±10	0.51 ±10
			Average	1.07±2.9%	0.509±3.5%
		Profile 025-035			
06-0936-94	6.32±2.2	13.0±2.4	6.69±1.2	1.06±2.5	0.513±2.7
		Profile 030-040			
06-0950-73	4.24±2.0	8.83±2.1	4.78±4.1	1.13±4.6	0.541±4.6
		Overall soil average		1.08 3.0%	0.512 3.6%

[a] All results are reported to a reference time of 1 January 1975 (001.000Z, 75).

1014406

Table 13. Wet-chemistry results of ^{241}Am vs Pu in Bikini vegetation.[a]

Master log number	Sample description	^{239}Pu dpm/g	$^{239+240}Pu$ dpm/g	^{241}Am dpm/g	$^{241}Am:^{239}Pu$ dpm ratio	$\frac{^{241}Am}{^{239+240}Pu}$ dpm ratio
01-0639-10	Old litter (coconut)	0.371 ± 4.7%	0.791 ± 4.5%	0.44 ±10%	1.2 ±11%	0.55 ±11%
01-0641-10	Composite litter: Papaya from 6 trees	0.729 ± 0.5	1.57 ± 1.3	0.75 ±19	1.0 ±19	0.48 ±19
01-0800-10	Mature pandanus leaves	0.139 ± 0.8	0.307 ± 1.4	0.165± 8.3	1.19± 8.3	0.537± 8.4
01-0802-10	Young pandanus leaves	———	0.14 ±18	0.073±14	———	0.53 ±23
01-0803-10	Pandanus roots	4.4 ±19	9.5 ±18	4.9 ±17	1.1 ±25	0.51 ±25
01-0804-10	Fallen fruit, papaya #2	0.0282± 1.3	0.0631± 3.2	0.032±10	1.1 ±11	0.50 ±11
01-0806-10	Young leaves, papaya #2	0.191 ± 4.2	0.426 ± 3.7	0.236± 7.1	1.24± 8.2	0.554± 8.0
01-0829-10	Root and crown of banana tree	0.393 ± 4.8	0.844 ± 6.3	0.43 ±11	1.1 ±12	0.50 ±12
01-0830-10	Breadfruit litter	0.0597± 3.4	0.139 ± 3.7	0.061±15	1.0 ±15	0.44 ±15
01-0839-10	Senescent leaves messerschmidia	0.0518± 5.5	0.118 ± 6.3	0.052±14	1.0 ±15	0.44 ±15
01-0847-10	Top litter Scaevola	0.121 ± 2.3	0.248 ± 3.8	0.127± 8.1	1.05± 8.4	0.512± 8.9
01-0850-10	Pandanus roots	1.43 ± 9.4	3.05 ± 9.2	1.67 ± 5.2	1.2 ±11	0.55 ±11
01-0853-10	Senescent Scaevola leaves	———	0.18 ±19	0.103± 8.7	———	0.56 ±21
				Average	1.11±8.1%	0.512±7.9%

[a] All results are reported to a reference time of 1 January 1975 (001.000Z, 75).

1014407

Table 14. Plutonium isotopic results from wet-chemistry analyses.[a]

Sample type	$^{240:239}Pu$ atom ratio	$^{241:239}Pu$ atom ratio	$^{240:239}Pu$ dpm ratio	$^{241:239}Pu$ dpm ratio
Bikini soil	0.305±5.2%	0.013±12%	1.14±5.2%	22±12%
Eneu soil	0.301±7.2	0.013±15	1.12±7.2	22±15
Bikini vegetation	0.315±7.6	0.014±14	1.18±7.6	24±14
Eneu vegetation	0.326±6.6	———	1.22±6.6	———

[a] All results are reported to a reference time of 1 January 1975 (001.000Z, 75).

Acknowledgments

The field portion of the June 1975 radiological survey of Bikini and Eneu Islands of Bikini Atoll was accomplished by a very intense and thorough effort by 21 people representing six different organizations. The number of samples collected and the amount of information obtained during the ten-day survey is a direct result of the cooperation and diligent effort of the following individuals:

Wayne Bliss
Environmental Protection Agency, Las Vegas, Nevada

Bruce Clegg
Lawrence Livermore Laboratory

Dave Coles
Lawrence Livermore Laboratory

Tom Crites
Lawrence Livermore Laboratory

Rod Eagle
Lawrence Livermore Laboratory

Harley Erwicker
Trust Territory of the Pacific Islands

Nat Greenhouse
Brookhaven National Laboratory

Paul Gudiksen
Lawrence Livermore Laboratory

Gale Holladay
Lawrence Livermore Laboratory

Bob Keller
Nevada Operations Office (ERDA)

Dennis McBreen
Trust Territory of the Pacific Islands

Tommy McCraw
Division of Operational Safety (ERDA)

Ben Mendoza
Lawrence Livermore Laboratory

Vic Nelson
University of Washington

1014408

Vic Noshkin
Lawrence Livermore Laboratory
Frank Reed
Environmental Protection Agency,
Las Vegas, Nevada
Jim Schweiger
Lawrence Livermore Laboratory
Robert Spies
Lawrence Livermore Laboratory
John Stewart
Nevada Operations Office (ERDA)
Marshall Stuart
Lawrence Livermore Laboratory

Thanks is extended to the following personnel for the superb job done in the processing of all soil and vegetation samples collected during the survey:

Jim Schweiger	LLL
Ben Mendoza	LLL
Greg Calvaird	RECO
Elizabeth Fletcher	RECO
Nancy Sawley	LLL
Cynthia Tafoya	LLL
Marshall Stuart	LLL

Gratitude is also extended to the following personnel from McClellan Central Laboratory for their excellent effort in the wet chemistry analysis of the soil vegetation and animal samples:

A. Ackland
R. Aduddel
L. Alexander
H. Aning
D. Beach
M. Beckinger
P. Carlson
W. Clark
R. Draper
W. Dunlap
D. Efurd
H. Erdman
D. Fletcher
W. Fuqua
C. Gay
J. Gholson
D. Griswold
R. Grogg
J. Hadl
H. Hamilton
R. Haslett
E. Henry
L. Hume
R. Jefferies
R. Johnson
M. Kantelo
P. Leciejewski
J. Lucas
R. Mayhew
C. McBrearty
G. Merrill
J. Miles
J. Miner
M. Mount
W. Myers
T. Nibarger
T. Opiela
R. Osborne

1014409

A. Paglione
J. Phelps
J. Phillips
C. Rheault
J. Riggs
R. San Miguel
B. Scholl
R. Schwarting
D. Seymour
J. Sexton
P. Sparman
W. Summers
W. Tracey
R. Wagoner
W. Washer
L. Williams
J. Wright

and to the following LLL personnel who were involved in the gamma spectrometry of all samples:

Jesse Meadows
Mike Allen
Ruth Anderson
Robert Wikkerink

William Phillips has done a fine job in establishing, writing, updating and correcting the rather extensive data bank generated by the analytical efforts.

The survey crew extends its thanks to Dr. Guy Haywood for medical support during the survey operation and to the Nevada Operations Office and Pacific Area Support Office for support services which resulted in a smooth and efficient survey. Support from the Kwajalein Missile Range and the site contractor, Global Associates, as well as from the crew of the R. V. Liktanur is greatly appreciated.

The outstanding cooperation of personnel from the Trust Territory of the Pacific Islands and from the Office of the District Administrator of the Marshall Islands, as well as that of the Bikini people, played an important part in the successful completion of the survey.

William L. Robison
Technical Director
Bikini Survey June 1975

BKI/gw

1014410

BLANK PAGE

1014411

Technical Information Department
LAWRENCE LIVERMORE LABORATORY
University of California | Livermore, California | 94550

1014412